

09/806002

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PATENT TRADEMARK OFFICE

CHAPTER II

TRANSMITTAL LETTER
TO THE UNITED STATES ELECTED OFFICE (EO/US)

(ENTRY INTO U.S. NATIONAL PHASE UNDER CHAPTER II)

PCT/DK99/00523	4 OCTOBER 1999	2 OCTOBER 1998
INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
A METHOD OF METALLIZING THE SURFACE OF A SOLID POLYMER SUBSTRATE AND THE PRODUCT OBTAINED		
TITLE OF INVENTION		

1. KRISTIAN GLEJBOL
2. BJORN WINTHER-JENSEN

APPLICANT(S)

Box PCT

Assistant Commissioner for Patents

Washington D.C. 20231

ATTENTION: EO/US

NOTE: The completion of those filing requirements that can be made at a time later than 30 months from the priority date results from the Commissioner exercising his judgment under the authority granted under 35 USC 371(d). The filing

CERTIFICATION UNDER 37 C.F.R. 1.10*

(Express Mail label number is **mandatory**.)

(Express Mail certification is optional.)

I hereby certify that this correspondence and the documents referred to as attached therein are being deposited with the United States Postal Service on this date March 23, 2001, in an envelope as "Express Mail Post Office to Addressee," Mailing Label Number EL728211834US, addressed to the: Assistant Commissioner for Patents, Washington, D.C. 20231.

MARIA MELIAN

(type or print name of person mailing paper)

Signature of person mailing paper

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receipt will show the actual date of receipt of the last item completing the entry into the national phase. See 37 C.F.R. §1.491 which states: "An international application enters the national state when the applicant has filed the documents and fees required by 35 USC 371(c) within the periods set forth in § 1.494 and § 1.495."

WARNING: Where the items are those which can be submitted to complete the entry of the international application into the national phase are subsequent to 30 months from the priority date the application is still considered to be in the international state and if mailing procedures are utilized to obtain a date the express mail procedure of 37 C.F.R. §1.10 must be used (since international application papers are not covered by an ordinary certificate of mailing - See 37 C.F.R. §1.8.

NOTE: Documents and fees must be clearly identified as a submission to enter the national state under 35 USC 371 otherwise the submission will be considered as being made under 35 USC 111. 37 C.F.R. § 1.494(f).

1. Applicant herewith submits to the United States Elected Office (EO/US) the following items under 35 U.S.C. 371:
 - a. ☒ This express request to immediately begin national examination procedures (35 U.S.C. 371(f)).
 - b. ☒ The U.S. National Fee (35 U.S.C. 371(c)(1)) and other fees (37 C.F.R. § 1.492) as indicated below:

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2.Fees

CLAIMS FEE	(1) FOR	(2) NUMBER FILED	(3) NUMBER EXTRA	(4) RATE	(5) CALCULATIONS
[]*	TOTAL CLAIMS	16 - 20 =		x \$ 18.00 =	\$
	INDEPENDENT CLAIMS	1 - 3 =		x \$ 80.00 =	
	MULTIPLE DEPENDENT CLAIM(S) (if applicable) + \$270.00				
BASIC FEE**	[] U.S. PTO WAS INTERNATIONAL PRELIMINARY EXAMINATION AUTHORITY Where an International preliminary examination fee as set forth in § 1.482 has been paid on the international application to the U.S. PTO: [] and the international preliminary examination report states that the criteria of novelty, inventive step (non-obviousness) and industrial activity, as defined in PCT Article 33(2) to (4) have been satisfied for all the claims presented in the application entering the national stage (37 CFR 1.492(a)(4)) \$100.00 [] and the above requirements are not met (37 CFR 1.492(a)(1)) \$690.00				
	[x] U.S. PTO WAS NOT INTERNATIONAL PRELIMINARY EXAMINATION AUTHORITY Where no international preliminary examination fee as set forth in § 1.482 has been paid to the U.S. PTO, and payment of an international search fee as set forth in § 1.445(a)(2) to the U.S. PTO: [] has been paid (37 CFR 1.492(a)(2)) \$710.00 [] has not been paid (37 CFR 1.492(a)(3)) \$1,000.00 [x] where a search report on the international application has been prepared by the European Patent Office or the Japanese Patent Office (37 CFR 1.492(a)(5)) \$860.00				
	Total of above Calculations				\$860.00
SMALL ENTITY	Reduction by ½ for filing by small entity, if applicable. Affidavit must be filed. (note 37 CFR 1.9, 1.27, 1.28)				-
	Subtotal				\$860.00
	Total National Fee				\$860.00
	Fee for recording the enclosed assignment document \$40.00 (37 CFR 1.21(h)). (See Item 13 below). See attached "ASSIGNMENT COVER SHEET".				
TOTAL	Total Fees enclosed				\$860.00

*See attached Preliminary Amendment Reducing the Number of Claims.

- A duplicate copy of this sheet is enclosed.

WARNING. If the translation of the international application and/or the oath or declaration have not been submitted by the applicant within thirty (30) months from the priority date, such requirements may be met within a time period set by the Office. 37 C.F.R. § 1.495(b)(2). The payment of the surcharge set forth in § 1.492(e) is required as a condition for accepting the oath or declaration later than thirty (30) months after the priority date. The payment of the processing fee set forth in § 1.492(f) is required for acceptance of an English translation later than thirty (30) months after the priority date. Failure to comply with these requirements will result in abandonment of the application. The provisions of § 1.136 apply to the period which is set. Notice of Jan. 3, 1993, 1147 O.G. 29 to 40.

- a. ☐ is transmitted herewith.
- b. ☐ is not required, as the application was filed with the United States Receiving Office.
- c. ☒ has been transmitted
- i. ☒ by the International Bureau.
- Date of mailing of the application (from form PCT/IB/308): 13 APRIL 2000.
- ii. ☐ by applicant on _____.
Date

- a. ☐ is transmitted herewith.
- b. ☐ is not required as the application was filed in English.
- c. ☐ was previously transmitted by applicant on _____
Date
- d. ☐ will follow.

5. ☒ Amendments to the claims of the International application under PCT Article 19 (35 U.S.C. 371(c)(3)):

NOTE: The Notice of January 7, 1993 points out that 37 C.F.R. § 1.495(a) was amended to clarify the existing and continuing practice that PCT Article 19 amendments must be submitted by 30 months from the priority date and this deadline may not be extended. The Notice further advises that: "The failure to do so will not result in loss of the subject matter of the PCT Article 19 amendments. Applicant may submit that subject matter in a preliminary amendment filed under section 1.121. In many cases, filing an amendment under section 1.121 is preferable since grammatical or idiomatic errors may be corrected." 1147 O.G. 29-40, at 36.

- a. ☐ are transmitted herewith.
- b. ☐ have been transmitted
- i. ☐ by the International Bureau.
Date of mailing of the amendment (from form PCT/IB/308): _____.
- ii. ☐ by applicant on _____
Date
- c. ☒ have not been transmitted as
- i. ☒ applicant chose not to make amendments under PCT Article 19.
Date of mailing of Search Report (from form PCT/ISA/210):
14 MARCH 2000.
- ii. ☐ the time limit for the submission of amendments has not yet expired.
The amendments or a statement that amendments have not been made will be transmitted before the expiration of the time limit under PCT Rule 46.1.
6. ☒ A translation of the amendments to the claims under PCT Article 19 (38 U.S.C. 371(c)(3)):
- a. ☐ is transmitted herewith.
- b. ☐ is not required as the amendments were made in the English language.
- c. ☒ has not been transmitted for reasons indicated at point 5(c) above.
7. ☒ A copy of the international examination report (PCT/IPEA/409)
- ☒ is transmitted herewith.
- ☐ is not required as the application was filed with the United States Receiving Office.
8. ☐ Annex(es) to the international preliminary examination report
- a. ☐ is/are transmitted herewith.
- b. ☐ is/are not required as the application was filed with the United States Receiving Office.
9. ☐ A translation of the annexes to the international preliminary examination report
- a. ☐ is transmitted herewith.
- b. ☐ is not required as the annexes are in the English language.

10. ☒ An oath or declaration of the inventor (35 U.S.C. 371(c)(4)) complying with 35 U.S.C. 115
- a. ☐ was previously submitted by applicant on _____.
Date
- b. ☐ is submitted herewith, and such oath or declaration
- i. ☐ is attached to the application.
- ii. ☐ identifies the application and any amendments under PCT Article 19 that were transmitted as stated in points 3(b) or 3(c) and 5(b); and states that they were reviewed by the inventor as required by 37 C.F.R. 1.70.
- c. ☒ will follow.

Other document(s) or information included:

11. ☒ An International Search Report (PCT/ISA/210) or Declaration under PCT Article 17(2)(a):
- a. ☐ is transmitted herewith.
- b. ☒ has been transmitted by the International Bureau.
Date of mailing (from form PCT/IB/308): 13 APRIL 2000.
- c. ☐ is not required, as the application was searched by the United States International Searching Authority.
- d. ☐ will be transmitted promptly upon request.
- e. ☐ has been submitted by applicant on _____.
Date
12. ☐ An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98:
- a. ☐ is transmitted herewith.
Also transmitted herewith is/are:
☐ Form PTO-1449 (PTO/SB/08A and 08B).
☐ Copies of citations listed.
- b. ☐ will be transmitted within THREE MONTHS of the date of submission of requirements under 35 U.S.C. 371(c).
- c. ☐ was previously submitted by applicant on _____.
Date
13. ☐ An assignment document is transmitted herewith for recording.

A separate ☐ "COVER SHEET FOR ASSIGNMENT (DOCUMENT) ACCOMPANYING NEW PATENT APPLICATION" or ☐ FORM PTO 1595 is also attached.

14. ☒ Additional documents:
- a. ☒ Copy of request (PCT/RO/101)
 - b. ☒ International Publication No. WO 00/20656
 - i. ☒ Specification, claims and drawing
 - ii. ☐ Front page only
 - c. ☐ Preliminary amendment (37 C.F.R. § 1.121)
 - d. ☒ Other

FORM PCT/IB/304; FORM PCT/IB/308; FORM PCT/IB/332;
FORM PCT/IPEA/416

15. ☒ The above checked items are being transmitted
- a. ☒ before 30 months from any claimed priority date.
 - b. ☐ after 30 months.
16. ☐ Certain requirements under 35 U.S.C. 371 were previously submitted by the applicant on _____, namely:
- _____
- _____
- _____

AUTHORIZATION TO CHARGE ADDITIONAL FEES

WARNING: *Accurately count claims, especially multiple dependent claims, to avoid unexpected high charges if extra claims are authorized.*

NOTE: *"A written request may be submitted in an application that is an authorization to treat any concurrent or future reply, requiring a petition for an extension of time under this paragraph for its timely submission, as incorporating a petition for extension of time for the appropriate length of time. An authorization to charge all required fees, fees under § 1.17, or all required extension of time fees will be treated as a constructive petition for an extension of time in any concurrent or future reply requiring a petition for an extension of time under this paragraph for its timely submission. Submission of the fee set forth in § 1.17(a) will also be treated as a constructive petition for an extension of time in any concurrent reply requiring a petition for an extension of time under this paragraph for its timely submission." 37 C.F.R. § 1.136(a)(3).*

NOTE: *"Amounts of twenty-five dollars or less will not be returned unless specifically requested within a reasonable time, nor will the payer be notified of such amounts; amounts over twenty-five dollars may be returned by check or, if requested, by credit to a deposit account." 37 C.F.R. § 1.26(a).*

- ☒ The Commissioner is hereby authorized to charge the following additional fees that may be required by this paper and during the entire pendency of this application to Account No. 12-0425.

☒ 37 C.F.R. 1.492(a)(1), (2), (3), and (4) (filing fees)

WARNING: *Because failure to pay the national fee within 30 months without extension (37 C.F.R. § 1.495(b)(2)) results in abandonment of the application, it would be best to always check the above box.*

☐ 37 C.F.R. 1.492(b), (c) and (d) (presentation of extra claims)

NOTE: *Because additional fees for excess or multiple dependent claims not paid on filing or on later presentation must*

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only be paid or these claims cancelled by amendment prior to the expiration of the time period set for response by the PTO in any notice of fee deficiency (37 C.F.R. § 1.492(d)), it might be best not to authorize the PTO to charge additional claim fees, except possible when dealing with amendments after final action.

- ☒ 37 C.F.R. 1.17 (application processing fees)
- ☒ 37 C.F.R. 1.17(a)(1)-(5)(extension fees pursuant to § 1.136(a).
- ☒ 37 C.F.R. 1.18 (issue fee at or before mailing of Notice of Allowance, pursuant to 37 C.F.R. 1.311(b))

NOTE: Where an authorization to charge the issue fee to a deposit account has been filed before the mailing of a Notice of Allowance, the issue fee will be automatically charged to the deposit account at the time of mailing the notice of allowance. 37 C.F.R. § 1.311(b).

NOTE: 37 C.F.R. 1.28(b) requires "Notification of any change in loss of entitlement to small entity status must be filed in the application . . . prior to paying, or at the time of paying . . . issue fee." From the wording of 37 C.F.R. § 1.28(b): (a) notification of change of status must be made even if the fee is paid as "other than a small entity" and (b) no notification is required if the change is to another small entity.

- ☐ 37 C.F.R. § 1.492(e) and (f) (surcharge fees for filing the declaration and/or filing an English translation of an International Application later than 30 months after the priority date).


SIGNATURE OF PRACTITIONER

WILLIAM R. EVANS

(type or print name of practitioner)

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05-10-01

JC07 Rec'd P07/PTO

09 MAY 2001

PCT



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PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: Kristian GLEJBOL, et al.

Serial No.: 09/806,002

Group No.:

Filed: March 23, 2001

Examiner:

For: METHOD OF METALLIZING THE SURFACE OF A SOLID POLYMER
SUBSTRATE AND THE PRODUCT OBTAINED

Attorney Docket No.: U 013327-7

Assistant Commissioner for Patents
Washington, D.C. 20231PRELIMINARY AMENDMENT

Please amend the above application as follows.

IN THE CLAIMS

4. (amended) A method according to claim 1, wherein step b) comprises treatment of the surface with a monomer vapour comprising 0.5 to 90 mole-%, preferably between 10 and 60 mole % of 2-ethyl cyano acrylate vapour.

CERTIFICATION UNDER 37 C.F.R. 1.10*(Express Mail label number is **mandatory**.)

(Express Mail certification is optional.)

I hereby certify that this correspondence and the documents referred to as attached therein are being deposited with the United States Postal Service on this date May 9, 2001, in an envelope as "Express Mail Post Office to Addressee", mailing Label Number EL723212786US, addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231.

MARIA MELIAN

(type or print name of person mailing paper)

Signature of person mailing paper

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6. (amended) A method according claim 1, wherein the polymer substrate is a polyolefine type, such as PE, PP, or an aryl type, such as styrene, a diene type, such as polybutadiene, polyisoprene, a silicone type, such as silicone rubber, a fluorine type, such as polytetrafluorethylene or its copolymers.

7. (amended) A method according to claim 1, wherein the polymer substrate is a PTFE PP.

8. (amended) A method according claim 1, wherein step a) comprises the generation of radicals b) use of a gas plasma generated by excitation of the gas in a direct current (DC), low frequency (LF), audio frequency (AF), radio frequency (RF) or microwave generated electric field.

9. (amended) A method according to claim 1, wherein the monomer pressure in step b) is between 0.1 and 100000 Pa.

10. (amended) A method according to claim 1, wherein the generation step a) is carried out for a period of between 0.01 and 1,000 seconds, and the treatment step b) is carried out for a period of between 0.1 and 1,000 seconds.

13. (amended) A method according to claim 1, wherein the temperature is the same under both step a) and step b), and preferably the temperature under both step a) and step b) is between 250 and 450 K.

14. (amended) A method according to claim 1, wherein the total pressure under step a) is equal to the total pressure under step b) and between 0.2 and 100000 Pa, more preferably between 0.2 and 10,000 Pa.

16. (amended) A polymer substrate metallized according to the method of claim 1.

Please add the following claims:

17. (new) A method according to claim 9, wherein the monomer pressure in step b) is between 10 and 1,000 Pa.

18. (new) The method according to claim 13, wherein the temperature is between 280 and 330 K.

19. (new) The method according to claim 14, wherein the total pressure is between 0.2 and 10,000 Pa.

20. (new) The method according to claim 19, wherein the total pressure is between 10 and 1,000 Pa.

Respectfully submitted,



William R. Evans
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Reg. No. 25858
Tel. No. (212) 708-1930

4. (amended) A method according to each of the preceding claims 1, 2 or 3, wherein step b) comprises treatment of the surface with a monomer vapour comprising 0.5 to 90 mole-%, preferably between 10 and 60 mole % of 2-ethyl cyano acrylate vapour.

6. (amended) A method according to each of the preceding claims 1, wherein the polymer substrate is a polyolefine type, such as PE, PP, or an aryl type, such as styrene, a diene type, such as polybutadiene, polyisoprene, a silicone type, such as silicone rubber, a fluorine type, such as polytetrafluorethylene or its copolymers.

7. (amended) A method according to each of the preceding claims 1-5, wherein the polymer substrate is a PTFE PP.

8. (amended) A method according to each of the preceding claims 1, wherein step a) comprises the generation of radicals b) use of a gas plasma generated by excitation of the gas in a direct current (DC), low frequency (LF), audio frequency (AF), radio frequency (RF) or microwave generated electric field.

9. (amended) A method according to each of the preceding claims 1, wherein the monomer pressure in step b) is between 0.1 and 100000 Pa, preferably between 10 and 1000 Pa.

10. (amended) A method according to each of the preceding claims 1, wherein the generation step a) is carried out for a period of between 0.01 and 10001,000 seconds, and the treatment step b) is carried out for a period of between 0.1 and 10001,000 seconds.

13. (amended) A method according to each of the preceding claims 1, wherein the temperature is the same under both step a) and step b), and preferably the temperature under both step a) and step b) is between 250 and 450 K, most preferably between 280 and 330 K.

14. (amended) A method according to each of the preceding claims 1, wherein the total pressure under step a) is equal to the total pressure under step b) the total pressure is preferably and between 0.2 and 100000 Pa, more preferably between 0.2 and 10000 Pa10, and most preferably between 10 and 1000000 Pa.

16. (amended) A polymer substrate metallized according to the method of each of the preceding claims 1-15. 1.

JC08 Rec'd PCT/PTO 23 MAR 2001

A method of metallizing the surface of a solid polymer substrate and the product obtained

- 5 The present invention relates to a method of metallizing the surface of polymer substrates such as polyolefins, fluorine containing polymers, polyalkylenes, polyarylenes and mixtures thereof and the product obtained.
- 10 Polymer surfaces with low surface energy such as untreated or unmodified polymer substrates are in general difficult to metallize using conventional methods based on electroless or galvanic plating methods.
- 15 One approach to obtain metallization on polymer surfaces is given in US 3,801,368, based on the fact that an electroless plating from a solution can be carried out, provided that the surface is first doped using noble metals like Au or Pt. The doping of the surface with noble met-
- 20 als prior to plating is, according to US 3,801,368, carried out by a vacuum evaporation or sputtering process. The advantage of using this type of process is that the energy of the metal-atoms hitting the surface often is sufficient to ensure a good bonding between the subse-
- 25 quently plated metal-film and the polymer surface.

The method described in US 3,801,368 is not universally applicable, but limited to certain polymers. Prior to the doping of the surface by noble metals, it is thus often

30 necessary to modify the chemical composition of the surface, in order to obtain sufficient adhesion between the subsequently plated film and the polymer surface.

The required change of chemical composition, or surface

35 treatment, depends on the substrate to be treated. Corona treatment is widely employed. The corona discharge con-

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sists of a number of small sparks created between the substrate surface and a counter electrode (a silent discharge). Most often this treatment is carried out in ambient atmosphere, and results in erosion of the surface as well as in a generation of free radicals due the energy density of the discharges and the reactivity of the atmosphere. One of the problems regarding this method is, that it is not suited for treatment prior to metallizing strongly curved surfaces or surfaces with many cavities. Other problems relate to the limited control over the resulting chemical composition of the surface.

Often a simple corona treatment is not sufficient and more specialised methods has to be employed to ensure a sufficient adhesion.

One possible method is described in WO 97/37844, where a method to obtain a coupling layer between the surface to be plated and the plated metal-film is described. According to WO 97/37844, the coupling layer is formed by vacuum deposition of a monomer on the surface, which is subsequently polymerised by an irradiation process. To ensure a good coupling between the surface and the coupling layer, and between the coupling layer and the plated metal film, plasma-treatment can be used. This process is especially suited for treatment of polymer films of infinite lengths, as the different processes (plasma-treatment, deposition of coupling layer and polymerisation of coupling layer) are carried out at different, separate, stations.

The nature of this process makes it furthermore not suitable for treatment of fluorine containing polymers like poly-tetra-fluor-ethylene (PTFE).

35

fluoro

A number of patents describe methods for electroless plating of PTFE. In JP 92-240189 a PTFE substrate is irradiated with UV laser in the presence of amine or amide followed by an immersing in an electroless metal plating bath. This results in a good bonding of the plated metal to the substrate. However, this method is very difficult to control and also very expensive.

US 3,956,535 discloses a further method of metallization where the object to be metallized is coated with a film having incorporated therein a hydrolysed metal complex. Upon this metal complex a metal or metal ion is adsorbed and acts as catalyst for autocatalytic metal deposition. This method is not usable on many types of polymer substrates due to poor adhesion between the coated film and the substrate.

Also US 4,952,286 describes a method which includes immersing an article to be plated into an electrolytic bath containing dissolved plating metal. Before immersing, the surface has been provided with areas of catalytic metal chalcogenide coating to be converted to a chemical resistant metal-coating.

US 4,057,663 describes a method to make hydrophobic polymers suitable for electroless plating using a concentrate containing P_2O_5 . This method is particularly suitable for polymers containing fluor, but not very useful for other polymers.

US 4,919,768 discloses another expensive method of metallization, where non-conducting surfaces are provided with a metal sulphide coating, acting as a base for direct electroplating.

Common to the above mentioned methods are, that they are either very expensive, complicated to use or not applicable to various types of polymer substrates.

- 5 The object of the present invention is to provide an industrially applicable method of binding a metal layer to a polymer substrate, which method does not comprise the above mentioned problems, resulting in good binding strength, and can be used on most polymer substrates.

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Another object of the present invention is to provide a fast method of binding a metal layer to a polymer surface, and thereby provide a good binding strength.

- 15 A further object of the present invention is to provide a method of metallizing a polymer surface, which does not result in any severe depolymerisation of the polymer substrate, in particular when the substrate material exhibits fluorine and/or tertiary carbon atoms, and by use of
20 which method the surface affinity against the metal is improved.

- A further object of the present invention is to provide a metallized polymer surface, which can be enhanced by
25 electrochemical deposition using any conventional method.

Another object of the present invention is to provide a method for metallizing a polymer substrate, which method is economical acceptable.

30

A further object of the present invention is to provide a method for metallizing a polymer substrate, which method is uncomplicated to use.

9 of an injection moulded polymer component

These objects are achieved by a method of metallizing a surface of a solid polymer substrate⁹ comprising the steps of

- 5 a) generating radicals on the substrate surface by subjecting it to a gas plasma,
- b) forming a layer on the surface using a plasma enhanced polymerisation process employing one or more monomers
- 10 comprising monomers selected among cyano acrylate, mono- and diacrylates, such as acrylic acid, triethylen glycol diacrylat, glycidyl acrylat, isocyanates, such as 1,4-diisocyanobutane, toluenediisocyanate, epoxy compounds, such as glycidyl methacrylate, preferably 2,3-epoxypropyl
- 15 methacrylat, allylic and vinylic compounds, such as vinyl acetic acid, vinyl norbonene, vinyl pyrrolidone, vinyl trimethoxysilane, vinyl trimethylsilane allylene, allyl alcohol, allyloxymethylsilane, allylphenol, allylurea 1-allyltheourea(thiosine-amine), vinyl-amine, vinyl-alcohol
- 20 or allyl-amine.
- c) providing a short surface deposition using a PVD or CVD process to deposit metal atoms, such as copper, tin, silver palladium, platinum, or gold
- 25 d) optionally providing a metallization of the surface by using a conventional electroless bath, or
- avoiding electroless metallization by using direct electrolytic metallization, when the metal layer formed in c)
- 30 has a thickness allowing electrolytic metallization.
- Step b) may start before step a) provided that step b) does not terminate until step a) is started, simultaneously with step a), under step a), or follow immediately
- 35 after step a). Step c) may start before step b), simulta-

neously with step b), follow immediately after step b) or within 8 months after step b), preferably within 6 months, and step d) may follow step c) or start simultaneously with step d).

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The method is preferably carried out in a reactor at least partly evacuated from air and water vapour. Further, the reactor should have a channel for feeding the working gas, and a channel for feeding the monomer or monomer mixture. The monomer or monomer mixture is introduced as a gas, e.g. by evaporation from a bottle.

Gas is fed into the reactor, and the plasma is generated (step a). Before, simultaneously or shortly thereafter monomer or monomer mixture is fed into the reactor (step b).

It is noted that the polymerisation preferably should be carried out at plasma discharge parameters resulting in a surface containing chemical groups promoting acid-base reactions with the subsequently deposited metal layer.

The generation step a) is preferably, as stated in claim 11, carried out for a period of between 0.01 and 1000 seconds, preferably between 1 and 500 seconds, more preferably between 10 and 60 seconds, and the treatment step b) is preferably carried out for a period of between 0.1 and 1000 seconds, preferably between 1 and 500 seconds, more preferably between 10 and 200 seconds.

The treatment step b) may continue when step a) has ended, even though there no longer are generated radicals on the substrate surface. This continuation of step b) will then result in a polymerisation of monomers onto the

new polymeric layer which already have been bound to the polymer surface.

If the surface of the polymer substrate is contaminated with water, oil or other contaminants, step a) is preferably carried out for more than 30 seconds, and step b) is started 10 to 30 seconds after step a) so as to clean the surface before the monomers are polymerised onto the substrate surface.

The partial pressure of the gas or the plasma in step a) is preferably between 0.1 and 10000 Pa.

The monomer pressure in step b) is preferably, as stated in claim 10, between 0.1 and 10000 Pa, more preferably between 10 and 1000 Pa.

The temperature is not important but should preferably, as stated in claim 14, be the same under both step a) and step b) and preferably the temperature under both step a) and step b) is between 250 and 450 K, most preferably between 280 and 330 K.

The total pressure, i.e. the sum of the partial pressures of the air, optionally water vapour, the gas or plasma and the monomer, under step a) is preferably, as stated in claim 15, equal to the total pressure under step b), the total pressure is preferably between 0.2 and 100000 Pa, more preferably between 0.2 and 10000 Pa, and most preferably between 10 and 1000 Pa.

The polymer substrate can be any polymer material provided that free radicals are created on the surface of the material when it is subjected to a gas plasma or other means of radical generation.

Preferably, as stated in claim 6, the polymer substrate is a polyolefine type, such as PE, PP, or an aryl type, such as styrene, a diene type, such as polybutadiene, polyisoprene, a silicone type, such as silicone rubber, a fluorine type, such as polytetrafluoroethylene or its copolymers.

The substrate can have any shape and any size, and may comprise complicated geometries. Preferably the polymer substrate is in the form of film, sheet, pipe, rod, porous or non-porous body, fabric, non-woven fabric, fibres or threads and/or as profiles made by extrusion, compression moulding, injection moulding, thermoforming or vacuum forming.

The generation of radicals on the substrate surface is preferably obtained by subjecting the substrate to a plasma discharge.

The plasma can be generated by any known methods, but preferably, as stated in claim 9, the gas plasma is generated by excitation of a gas in a direct current (DC), low frequency (LF), radio frequency (RF) or microwave (MW) generated electric field. Most preferably the gas plasma is generated by excitation of a gas in a direct current (DC) or by excitation using low frequency (LF).

The intensity of the used gas plasma should preferably have a level ensuring creation of radicals in the polymer surface. If the level is too high, this may result in severe damage of the bulk-polymer (depolymerisation). Hence, the power level of the plasma should be optimised so that surface radicals are created, but no serious damage is made to the bulk.

A preferred method of generating a plasma discharge is described in the applicant's European patent application No. EP 96610018.2.

- 5 Preferred embodiments for the invention are specified in claims 2 - 15.

10 *Experimental systems*

- Although it is possible to carry out all necessary steps in a single vacuum vessel, initial experiments were carried out in two separate systems. In one of the systems the virgin polymer surface was altered by the use of a plasma treatment using a working gas and monomers. In the other vacuum system metal atoms were subsequently deposited to the surface of the test samples. Although possible in an industrial production, the approach of using separate machines for the two steps is highly inefficient. The results obtained from these experiments do, however, reflect the results obtainable if a custom made system, able to perform both tasks is used.
- 25 The first step was on all samples carried out in an experimental plasma system, essentially consisting of a 22 litre vacuum vessel having two inlet channels for gas and for monomer, respectively, and an electrode arrangement as described above with reference to the applicant's EP application No. 96610018.2 The plasma is generated using the 3-phase technique described above with a frequency of 50 Hz and U_0 of 240 to 280V. 27 electrodes are arranged in a circle around the vacuum vessel with a diameter of 17 cm. Every third of the electrodes is connected to U_r , another every third of the electrodes is connected to U_s ,

and the last every third of the electrodes is connected to U_t .

5 The second step was on all samples carried out in an experimental plasma system, essentially consisting of a glass bowl having an internal volume of 15 litres. To atomise the metal and transfer it from the target to the sample surface a custom made magnetron was constructed. The magnetron is essentially a copper plate, having a diameter of 10 cm and a thickness of 4 mm. The back-side of the copper plate is shielded using a ceramic container. On the backside of the copper-plate a magnet is mounted. The magnetron is operated by applying a negative DC potential of 450 V. During operation a current of 0.05 A is drawn between the magnetron and the positive parts of the chamber. During all experiments argon is used as the working gas for the magnetron.

20 Both plasma systems are pumped using the same vacuum system comprising an Edwards EH 250 Roots-blower, backed by an Alcatel rotary vane pump. The pressure is monitored using an Alcatel pirani-gauge, mounted on the start of the pumping-line.

25 During step a) and b) the sample is held at floating potential.

Example 1

30 Covering a 100x50x3 mm sheet of polytetrafluorethylene (PTFE) with a conductive layer of metal.

Step 1

35 The sheet was subjected to an argon plasma discharge, generated as described above, for 20 s at a pressure of

0.1 mbar. Hereafter a vapour essentially comprising of 2-ethyl cyano acrylate and glycidylmetacrylate was let into the argon plasma at a pressure of 0.15 mbar for 30 s.

5 Step 2

After this treatment 100 nm of silver (Ag) was deposited onto the surface, making the surface conductive. The deposition was accomplished by using a silver sheet as a target on the magnetron, and the magnetron was operated
10 for 60 seconds.

Step 3

After Ag deposition, the sheet was immersed in an electroless copper bath, ("Shipley Cuposit 251 electroless copper"). Due to the combined treatment in step 1 and 2
15 an adherent copper coating was formed where Ag was deposited.

The copper layer can be 0.5 μm or more, depending on the
20 treatment time in the electroless copper bath. The average thickness obtained after a given time is as claimed by Shipley 2.5 μm per hours.

The adhesion strength of the copper layer was examined by
25 the standard well known to people skilled in the art, DIN 53 151 having a scale from GT 0 to GT 5, where zero is excellent. The treated sample in this example complied the standard and got the score Gt 0 - Gt 1.

30 Example 2

Covering of a component made from polypropylene (PP) with a shielding metallic layer.

Step 1

The component was subjected to an argon plasma as described above for 10 s at a pressure of 1 mbar. Hereafter a vapour essentially comprising of toluene-2,4-diisocyanate was let into the plasma chamber at a pressure of 2 mbar for 20 s.

Step 2

After this treatment 2 ML (monolayers) of platinum (Pt) was accomplished by using a Pt sheet as a target on the magnetron, and the magnetron was operated for 5 seconds.

Step 3

The component was immersed in an electroless copper bath, ("Shipley Cuposit 251 electroless copper"). Due to the combined treatment in steps 1 and 2 an adherent copper coating was formed where Pt was deposited on the PP component.

20 Example 3

Covering a PTFE surface with a 0.5 μm metal layer.

Steps 1 and 2 were conducted as described in example 2.

25

Step 3

The component was immersed in an electroless copper bath, ("Shipley Cuposit 251 electroless copper"). Due to the combined treatment in steps 1 and 2 an adherent copper coating was formed where Ag was deposited.

Example 4a

35 Covering a PTFE surface with a 5 μm metal layer.

Steps 1 - 2 identical to the steps in example 2.

Step 3

Copper was then deposited on the Ag covered surface using
5 a conventional electroplating technique.

Example 4b

Covering a PTFE surface with a 5 μm metal layer

10

Steps 1 - 3 identical to the steps in example 3

Step 4

More copper was then deposited on the surface using a
15 conventional electroplating technique.

9 of an injection moulded polymer
CLAIMS. component

1. A method of metallizing a solid polymer substrate^φ comprising the steps of

- 5
- a) generating radicals on the substrate surface by subjecting it to a gas plasma,
- b) forming a layer on the surface using a plasma enhanced polymerisation process employing one or more monomers comprising monomers selected among cyano acrylate, mono- and diacrylates, such as acrylic acid, triethylen glycol diacrylate, glycidyl acrylate, isocyanates, ~~such as~~ 1,4-diisocyanobutane, toluenediisocyanate, epoxy compounds, 15 such as glycidyl methacrylate, preferably 2,3-epoxypropyl methacrylate, allylic and vinylic compounds, ~~such as~~ vinyl acetic acid, vinyl norbornene, vinyl pyrrolidone, vinyl trimethoxysilane, vinyl trimethylsilane allylene, allyl alcohol, allyloxymethylsilane, allylphenol, allylurea 20 1-allylthiourea (thiosineamine),
- c) providing a short surface deposition using a PVD or CVD process to deposit metal atoms, such as copper, tin, silver palladium, platinum, or gold, and 25
- d) optionally providing a metallization of the surface by using a conventional electroless bath, or
- avoiding electroless metallization by using direct electrolytic metallization, when the metal layer formed in c) 30 has a thickness allowing electrolytic metallization.

2. A method according to claim 1, wherein the (catalytic metal) comprises Pt, Ag, Pd, Cu and Au.

35

2 x + — preferably selected from the group
consisting of
(-) metal layer

3. A method according to claim 1, wherein the monomer or monomer mixture comprises one or more of cyanoacrylate and glycidyl metacrylate, preferably 2,3-epoxypropyl methacrylate.

5

4. A method according to each of the preceding claims 1, 2 or 3, wherein step b) comprises treatment of the surface with a monomer vapour comprising 0.5 to 90 mole-%, preferably between 10 and 60 mole % of 2-ethyl cyano acrylate vapour.

10

5. A method according to claim 4, wherein the monomer prior to the vaporisation consists essentially of 2-ethyl cyano acrylate, an acid having a partial vapour pressure in the plasma which is lower than the partial vapour pressure of 2-ethyl cyano-acrylate, and up to 40 weight-% of another filler, preferably an acid having a partial vapour pressure in the plasma which is lower than half the partial vapour pressure of 2-ethyl cyano-acrylate, most preferably the acid is a polyphosphoric acid and is present prior to the vaporisation in a concentration up to 10 weight-%.

15

20

6. A method according to each of the preceding claims, wherein the polymer substrate is a polyolefine type, such as PE, PP, or an aryl type, such as styrene, a diene type, such as polybutadiene, polyisoprene, a silicone type, such as silicone rubber, a fluorine type, such as polytetrafluorethylene or its copolymers.

30

7. A method according to each of the preceding claims 1-5, wherein the polymer substrate is a PTFE (and) PP. (-) or

~~8. A method according to each of the preceding claims, wherein the polymer substrate is an injection moulded~~ > ok

35

~~polymer component, a polymer fibre, a polymer thread or a polymer filler.~~ 7 cl

- 8 9. A method according to each of the preceding claims,
5 wherein step a) comprises the generation of radicals by use of a gas plasma generated by excitation of the gas in a direct current (DC), low frequency (LF), audio frequency (AF), radio frequency (RF) or microwave generated electric field.
- 10 9 10. A method according to each of the preceding claims,
wherein the monomer pressure in step b) is between 0.1 and 10000 Pa, preferably between 10 and 1000 Pa.
- 10 11. A method according to each of the preceding claims,
15 wherein the generation step a) is carried out for a period of between 0.01 and 1000 seconds, and the treatment step b) is carried out for a period of between 0.1 and 1000 seconds.
- 20 11 12. A method according to claim 11, wherein step a) is carried out for more than 30 seconds, and step b) is started 10 to 30 seconds after step a).
- 10 12 13. A method according claim 11, wherein the generation
25 step a) is carried out for a period of between 10 and 60 seconds, and the treatment step b) is carried out for a period of between 10 and 200 seconds.
- 13 14. A method according to each of the preceding claims,
30 wherein the temperature is the same under both step a) and step b), and preferably the temperature under both step a) and step b) is between 250 and 450 K, most preferably between 280 and 330 K.

14 15. A method according to each of the preceding claims,
wherein the total pressure under step a) is equal to the
total pressure under step b), the total pressure is pref-
erably between 0.2 and 100000 Pa, more preferably between
5 0.2 and 10000 Pa, and most preferably between 10 and 1000
Pa.

15 16. A method according to claim 1, where step b) starts
before step a) provided that step b) does not terminate
10 until step a) is started, simultaneously with step a),
under step a), or follows immediately after step a),
where step c) starts before step b), simultaneously with
step b), follows immediately after step b) or within 6
months after step b), preferably within 6 months, and
15 where step d) follows step c) or starts simultaneously
with step d).

16 17. A polymer substrate metallized according to the
method of each of the preceding claims 1-16.

20

15

A method of metallizing the surface of a solid polymer substrate and the product obtained

ABSTRACT

5

A method of metallizing a solid polymer substrate comprising the steps of

10 a) generating radicals on the substrate surface by subjecting it to a gas plasma,

b) forming a layer on the surface using a plasma enhanced polymerisation process employing one or more monomers comprising monomers selected among cyano acrylate, mono-
15 and diacrylates, such as acrylic acid, triethylen glycol diacrylate, glycidyl acrylate, isocyanates, such as 1,4-diisocyanobutane, toluenediisocyanate, epoxy compounds, such as glycidyl methacrylate, preferably 2,3-epoxypropyl methacrylate, allylic and vinylic compounds, such as vi-
20 nyl acetic acid, vinyl norbonene, vinyl pyrrolidone, vinyl trimethoxysilane, vinyl trimethylsilane allylene, allyl alcohol, allyloxymethylsilane, allylphenol, allylurea 1-allyltheourea(thiosineamine),

25 c) providing a short surface deposition using a PVD or CVD process to deposit metal atoms, such as copper, tin, silver palladium, platinum, or gold, and

30 d) optionally providing a metallization of the surface by using a conventional electroless bath, or

avoiding electroless metallization by using direct electrolytic metallization, when the metal layer formed in c) has a thickness allowing electrolytic metallization.

35

Practitioner's Docket No. _____

PATENT

COMBINED DECLARATION AND POWER OF ATTORNEY(ORIGINAL, DESIGN, NATIONAL STAGE OF PCT, SUPPLEMENTAL, DIVISIONAL,
CONTINUATION, OR C-I-P)

As a below named inventor, I hereby declare that:

TYPE OF DECLARATION

This declaration is of the following type:

(check one applicable item below)

- ☐ original.
☐ design.
☐ supplemental.

NOTE: If the declaration is for an International Application being filed as a divisional, continuation or continuation-in-part application, do not check next item; check appropriate one of last three items.

- ☒ national stage of PCT.

NOTE: If one of the following 3 items apply, then complete and also attach ADDED PAGES FOR DIVISIONAL, CONTINUATION OR C-I-P.

NOTE: See 37 C.F.R. § 1.63(d) (continued prosecution application) for use of a prior nonprovisional application declaration in the continuation or divisional application being filed on behalf of the same or fewer of the inventors named in the prior application.

- ☐ divisional.
☐ continuation.

NOTE: Where an application discloses and claims subject matter not disclosed in the prior application, or a continuation or divisional application names an inventor not named in the prior application, a continuation-in-part application must be filed under 37 C.F.R. § 1.53(b) (application filing requirements-nonprovisional application).

- ☐ continuation-in-part (C-I-P).

INVENTORSHIP IDENTIFICATION

WARNING: If the inventors are each not the inventors of all the claims, an explanation of the facts, including the ownership of all the claims at the time the last claimed invention was made, should be submitted.

My residence, post office address and citizenship are as stated below, next to my name. I believe that I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter that is claimed, and for which a patent is sought on the invention entitled:

A method of metallizing the surface of a solid polymer substrate and the product obtained.

TITLE OF INVENTION
A method of metallizing the surface of a solid polymer
substrate and the product obtained.

SPECIFICATION IDENTIFICATION

The specification of which:

(complete (a), (b), or (c))

(a) ☐ is attached hereto.

NOTE: "The following combinations of information supplied in an oath or declaration filed on the application filing date with a specification are acceptable as minimums for identifying a specification and compliance with any one of the items below will be accepted as complying with the identification requirement of 37 C.F.R. § 1.63:

"(1) name of inventor(s), and reference to an attached specification which is both attached to the oath or declaration at the time of execution and submitted with the oath or declaration on filing;

"(2) name of inventor(s), and attorney docket number which was on the specification as filed; or

"(3) name of inventor(s), and title which was on the specification as filed."

Notice of July 13, 1995 (1177 O.G. 60).

(b) ☐ was filed on _____, ☐ as Application No. _____
☐ and was amended on _____ (if applicable).

NOTE: Amendments filed after the original papers are deposited with the PTO that contain new matter are not accorded a filing date by being referred to in the declaration. Accordingly, the amendments involved are those filed with the application papers or, in the case of a supplemental declaration, are those amendments claiming matter not encompassed in the original statement of invention or claims. See 37 C.F.R. § 1.67.

NOTE: "The following combinations of information supplied in an oath or declaration filed after the filing date are acceptable as minimums for identifying a specification and compliance with any one of the items below will be accepted as complying with the identification requirement of 37 C.F.R. § 1.63:

"(1) name of inventor(s), and application number (consisting of the series code and the serial number; e.g., 08/123,456);

"(2) name of inventor(s), serial number and filing date;

"(3) name of inventor(s) and attorney docket number which was on the specification as filed;

"(4) name of inventor(s), title which was on the specification as filed and filing date;

"(5) name of inventor(s), title which was on the specification as filed and reference to an attached specification which is both attached to the oath or declaration at the time of execution and submitted with the oath or declaration; or

"(6) name of inventor(s), title which was on the specification as filed and accompanied by a cover letter accurately identifying the application for which it was intended by either the application number (consisting of the series code and the serial number; e.g., 08/123,456), or serial number and filing date. Absent any statement(s) to the contrary, it will be presumed that the application filed in the PTO is the application which the inventor(s) executed by signing the oath or declaration."

Notice of July 13, 1995 (1177 O.G. 60), M.P.E.P. § 601(a), 6th ed., rev.3.

- (c) ☒ was described and claimed in PCT International Application No. PCT/DK99/00523 filed on Oct. 4, 1999 and as amended under PCT Article 19 on _____ (if any).

SUPPLEMENTAL DECLARATION (37 C.F.R. § 1.67(b))

(complete the following where a supplemental declaration is being submitted)

☐ I hereby declare that the subject matter of the

☐ attached amendment

☐ amendment filed on _____.

was part of my/our invention and was invented before the filing date of the original application, above identified, for such invention.

ACKNOWLEDGMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information, which is material to patentability as defined in 37, Code of Federal Regulations, § 1.56,

(also check the following items, if desired)

☐ and which is material to the examination of this application, namely, information where there is a substantial likelihood that a reasonable Examiner would consider it important in deciding whether to allow the application to issue as a patent, and

☐ in compliance with this duty, there is attached an information disclosure statement, in accordance with 37 C.F.R. § 1.98.

PRIORITY CLAIM (35 U.S.C. § 119(a)-(d))

NOTE: "The claim to priority need be in no special form and may be made by the attorney or agent if the foreign application is referred to in the oath or declaration as required by § 1.63. The claim for priority and the certified copy of the foreign application specified in 35 U.S.C. § 119(b) must be filed in the case of an interference (§ 1.630), when necessary to overcome the date of a reference relied upon by the examiner, when specifically required by the examiner, and in all other situations, before the patent is granted. If the claim for priority or the certified copy of the foreign application is filed after the date the issue fee is paid, it must be accompanied by a petition requesting entry and by the fee set forth in § 1.17(i). If the certified copy is not in the English language, a translation need not be filed except in the case of interference; or when necessary to overcome the date of a reference relied upon by the examiner; or when specifically required by the examiner, in which event an English language translation must be filed together with a statement that the translation of the certified copy is accurate." 37 C.F.R. § 1.55(a).

I hereby claim foreign priority benefits under Title 35, United States Code, § 119(a)-(d) of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

(complete (d) or (e))

- (d) ☐ no such applications have been filed.
(e) ☒ such applications have been filed as follows.

NOTE: Where item (c) is entered above and the International Application which designated the U.S. itself claimed priority check item (e), enter the details below and make the priority claim.

**PRIOR FOREIGN/PCT APPLICATION(S) FILED WITHIN 12 MONTHS
(6 MONTHS FOR DESIGN) PRIOR TO THIS APPLICATION
AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. § 119(a)-(d)**

COUNTRY (OR INDICATE IF PCT)	APPLICATION NUMBER	DATE OF FILING DAY, MONTH, YEAR	PRIORITY CLAIMED UNDER 35 USC 119
Denmark	PA 1998 01247	04 Oct. 1998	<input checked="" type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO

CLAIM FOR BENEFIT OF PRIOR U.S. PROVISIONAL APPLICATION(S)
(35 U.S.C. § 119(e))

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below:

PROVISIONAL APPLICATION NUMBER

FILING DATE

_____/_____
_____/_____
_____/_____

CLAIM FOR BENEFIT OF EARLIER U.S./PCT APPLICATION(S)
UNDER 35 U.S.C. § 120

- [] The claim for the benefit of any such applications are set forth in the attached
ADDED PAGES TO COMBINED DECLARATION AND POWER OF
ATTORNEY FOR DIVISIONAL, CONTINUATION OR CONTINUATION-
IN-PART (C-I-P) APPLICATION.

ALL FOREIGN APPLICATION(S), IF ANY, FILED MORE THAN 12 MONTHS
(6 MONTHS FOR DESIGN) PRIOR TO THIS U.S. APPLICATION

NOTE: If the application filed more than 12 months from the filing date of this application is a PCT filing forming the basis for this application entering the United States as (1) the national stage, or (2) a continuation, divisional, or continuation-in-part, then also complete ADDED PAGES TO COMBINED DECLARATION AND POWER OF ATTORNEY FOR DIVISIONAL, CONTINUATION OR C-I-P APPLICATION for benefit of the prior U.S. or PCT application(s) under 35 U.S.C. § 120.

POWER OF ATTORNEY

I hereby appoint the following practitioner(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

(list name and registration number)

JOSEPH H. HANDELMAN, 26179

JOHN RICHARDS, 31053

RICHARD J. STREIT, 25765

PETER D. GALLOWAY, 27885

IAN C. BAILLIE, 24090

THOMAS F. PETERSON, 24790

RICHARD P. BERG, 28145

JULIAN H. COHEN, 20302

WILLIAM R. EVANS, 25858

JANET I. CORD, 33778

CLIFFORD J. MASS, 30086

CYNTHIA R. MILLER, 34678

12

(Check the following item, if applicable)

- [] I hereby appoint the practitioner(s) associated with the Customer Number provided below to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.
- [] Attached, as part of this declaration and power of attorney, is the authorization of the above-named practitioner(s) to accept and follow instructions from my representative(s).

SEND CORRESPONDENCE TO

DIRECT TELEPHONE CALLS TO:
(Name and telephone number)

Ladas & Parry
26 West 61st Street
New York, N.Y. 10023

DECLARATION

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

SIGNATURE(S)

NOTE: Carefully indicate the family (or last) name, as it should appear on the filing receipt and all other document.

NOTE: Each inventor must be identified by full name, including the family name, and at least one given name without abbreviation together with any other given name or initial, and by his/her residence, post office address and country of citizenship. 37 C.F.R. § 1.63(a)(3).

NOTE: Inventors may execute separate declarations/oaths provided each declaration/oath sets forth all the inventors. Section 1.63(a)(3) requires that a declaration/oath, inter alia, identify each inventor and prohibits the execution of separate declarations/oaths which each sets forth only the name of the executing inventor. 62 Fed. Reg. 53,131, 53,142, October 10, 1997.

Full name of sole or first inventor

Kristian
(Given Name)

1-10
(Middle Initial or Name)

GLEJBØL
Family (Or Last Name)

Inventor's signature (X) Kristian Gjelbøl

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WINTER-JENSEN
Family (Or Last Name)

Inventor's signature Bjørn Winter-Jensen

Date 25/04-01 Country of Citizenship Denmark

Residence Hornemannsgade 17, DK-2100 Copenhagen Ø.

Post Office Address Same

Full name of third joint inventor, if any

(Given Name)

(Middle Initial or Name)

Family (Or Last Name)

Inventor's signature

Date Country of Citizenship

Residence

Post Office Address

(check proper box(es) for any of the following added page(s)
that form a part of this declaration)

- ☐ **Signature** for fourth and subsequent joint inventors. *Number of pages added* _____

* * *

- ☐ **Signature** by administrator(trix), executor(trix) or legal representative for deceased or incapacitated inventor. *Number of pages added* _____

* * *

- ☐ **Signature** for inventor who refuses to sign or cannot be reached by person authorized under 37 C.F.R. § 1.47. *Number of pages added* _____

* * *

- ☐ Added page for **signature** by one joint inventor on behalf of deceased inventor(s) where legal representative cannot be appointed in time. (37 C.F.R. § 1.47)

* * *

- ☐ Added pages to combined declaration and power of attorney for divisional, continuation, or continuation-in-part (C-I-P) application.

☐ Number of pages added _____

* * *

- ☐ Authorization of practitioner(s) to accept and follow instructions from representative.

(If no further pages form a part of this Declaration,
then end this Declaration with this page and check the following item)

☒ This declaration ends with this page.